

Europäisches Patentamt  
European Patent Office  
Office européen des brevets



(11) **EP 0 741 970 A1**

(12)

**EUROPEAN PATENT APPLICATION**

published in accordance with Art. 158(3) EPC

(43) Date of publication:  
13.11.1996 Bulletin 1996/46

(21) Application number: 95902287.2

(22) Date of filing: 01.12.1994

(51) Int. Cl.<sup>6</sup>: **A01N 37/50**, A01N 37/22,  
A01N 37/32, A01N 37/34,  
A01N 43/08, A01N 43/36,  
A01N 43/40, A01N 43/50,  
A01N 43/54, A01N 43/76,  
A01N 43/78

(86) International application number:  
PCT/JP94/02023

(87) International publication number:  
WO 95/15083 (08.06.1995 Gazette 1995/24)

(84) Designated Contracting States:  
ES FR IT

(30) Priority: 02.12.1993 JP 303143/93  
27.12.1993 JP 333787/93  
19.04.1994 JP 80219/94  
21.04.1994 JP 83005/94  
13.05.1994 JP 99785/94  
16.05.1994 JP 100833/94  
25.05.1994 JP 111298/94  
30.05.1994 JP 116321/94  
15.07.1994 JP 164180/94  
15.07.1994 JP 164183/94

(71) Applicant: **SUMITOMO CHEMICAL COMPANY LIMITED**  
Osaka-shi, Osaka 541 (JP)

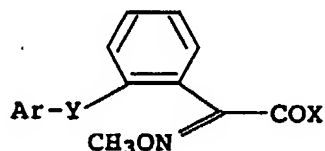
(72) Inventor: **OGURI, Yukio**  
Toyonaka-shi, Osaka 561 (JP)

(74) Representative: **Brewster, Andrea Ruth et al**  
**MEWBURN ELLIS**  
York House  
23 Kingsway  
London WC2B 6HP (GB)

(54) **BACTERICIDAL COMPOSITION**

(57) The present invention provides a fungicidal composition characterized by containing as active ingredients

(a) a compound represented by the formula I:



(wherein Ar is a substituted or unsubstituted phenyl group, Y is an oxygen atom, an oxymethylene group or a methyleneoxy group, X is NR<sub>1</sub>R<sub>2</sub> or OR<sub>3</sub>, and R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub>, which may be the same or different, are hydrogen atoms or alkyl groups having 1 to 4 carbon atoms), and

(b) at least one compound selected from the group consisting of ethylenebis(dithiocarbamate) type fungicidal compounds, N-(3,5-dichlorophenyl)imide type fungicidal compounds, Chlorothalonil, phthalimide type fungicidal compounds, anilide type fungicidal compounds, Cymoxanil, Fosetyl, cyanopyrrole type fungicidal compounds, anilino type heterocyclic fungicidal compounds, benzimidazole type fungicidal compounds and their precursors, sulfur, copper compounds, and carbamate type fungicidal compounds for controlling fungi tolerant to benzimidazole type fungicides.

EP 0 741 970 A1

## Description

## TECHNICAL FIELD

5 The present invention relates to a fungicidal composition mainly for agricultural and horticultural use.

## BACKGROUND ART

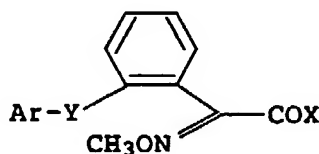
10 Various agricultural and horticultural fungicides have been known, but there are a great variety of diseases to be controlled and it is difficult to specify the kind of the disease in practice and control the disease by choosing a fungicide suitable for the disease. In addition, it is necessary to cope with the appearance of fungi which have acquired tolerance to specific agents and the occurrence of novel diseases due to the change of the mode of agriculture. For these reasons and the like, there is desired a fungicidal composition having a high activity and a wide antimicrobial spectrum.

15 The present invention provides a fungicidal composition which has a high activity and a wide antimicrobial spectrum and is markedly effective also against diseases caused by plant pathogenic fungi which have acquired tolerance to certain fungicides.

## DISCLOSURE OF THE INVENTION

20 In detail, the present invention provides a fungicidal composition characterized by containing as active ingredients

(a) a compound represented by the formula I:



(wherein Ar is a substituted or unsubstituted phenyl group, Y is an oxygen atom, an oxymethylene group or a methyleneoxy group, X is  $\text{NR}_1\text{R}_2$  or  $\text{OR}_3$ , and  $\text{R}_1$ ,  $\text{R}_2$  and  $\text{R}_3$ , which may be the same or different, are hydrogen atoms or alkyl groups having 1 to 4 carbon atoms), and

35 (b) at least one compound selected from the group consisting of ethylenebis(dithiocarbamate) type fungicidal compounds, N-(3,5-dichlorophenyl)imide type fungicidal compounds, Chlorothalonil, phthalimide type fungicidal compounds, anilide type fungicidal compounds, Cymoxanil, Fosetyl, cyanopyrrole type fungicidal compounds, anilino heterocyclic fungicidal compounds, benzimidazole type fungicidal compounds and their precursors, sulfur, copper compounds, and carbamate type fungicidal compounds for controlling fungi tolerant to benzimidazole type fungicides.

The compound of the formula I used in the present invention includes the compounds disclosed in Japanese Patent Unexamined Publication Nos. 63-23852, 3-246268 and 4-288045, etc.

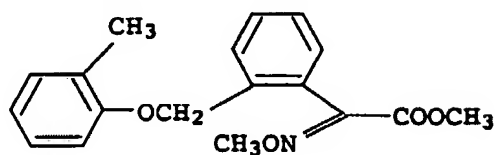
45 In the formula I, the substituted or unsubstituted phenyl group represented by Ar refers to a phenyl group which may be substituted by one or more substituents selected from alkyl groups having 1 to 4 carbon atoms (e.g. methyl group and ethyl group), halogen atoms (e.g. chlorine atom and bromine atom), haloalkyl groups having 1 to 4 carbon atoms (e.g. trifluoromethyl group), alkoxy groups having 1 to 4 carbon atoms (e.g. methoxy group), haloalkoxy groups (e.g. trifluoromethoxy group), cyano group, etc.

50 Y is an oxygen atom, an oxymethylene group or a methyleneoxy group, X is  $\text{NR}_1\text{R}_2$  or  $\text{OR}_3$ , and  $\text{R}_1$ ,  $\text{R}_2$  and  $\text{R}_3$ , which may be the same or different, are hydrogen atoms or alkyl groups having 1 to 4 carbon atoms, such as methyl groups.

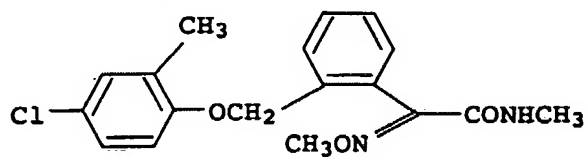
Some preferable specific examples of the compound of the formula I are given below:

55

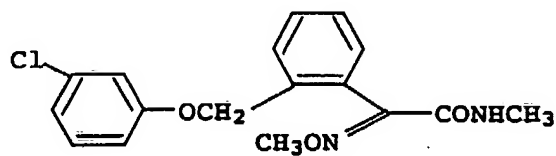
(la) methyl  $\alpha$ -methoxyimino-2-[(2-methylphenoxy)methyl]phenylacetate



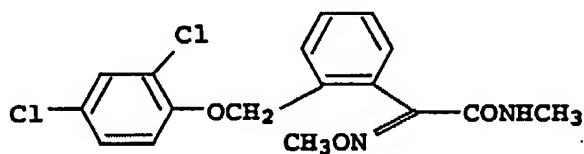
(lb) N-methyl- $\alpha$ -methoxyimino-2-[(4-chloro-2-methylphenoxy)methyl]phenylacetamide



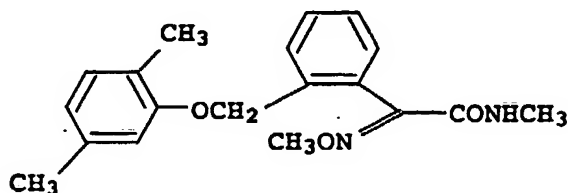
(lc) N-methyl- $\alpha$ -methoxyimino-2-[(3-chlorophenoxy)methyl]phenylacetamide



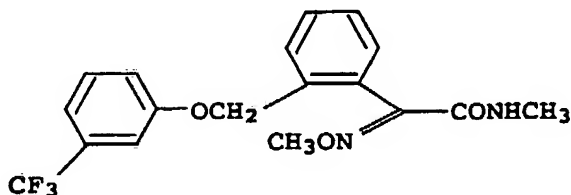
(ld) N-methyl- $\alpha$ -methoxyimino-2-[(2,4-dichlorophenoxy)methyl]phenylacetamide



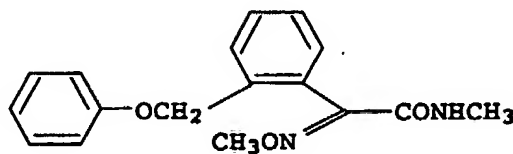
(Ie) N-methyl- $\alpha$ -methoxyimino-2-[(2,5-dimethylphenoxy)methyl]phenylacetamide



(If) N-methyl- $\alpha$ -methoxyimino-2-[(3-trifluoromethylphenoxy)methyl]phenylacetamide



(Ig) N-methyl- $\alpha$ -methoxyimino-2-phenoxyphenylacetamide

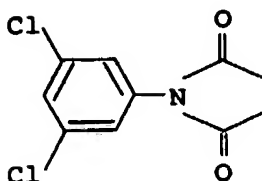


The fungicidal compound(s) used in admixture with the above-mentioned compound is (b) at least one compound selected from the group consisting of ethylenebis(dithiocarbamate) type fungicidal compounds, N-(3,5-dichlorophenyl)imide type fungicidal compounds, Chlorothalonil (tetrachloroisophthalonitrile), phthalimide type fungicidal compounds, anilide type fungicidal compounds, Cymoxanil, Fosetyl, cyanopyrrole type fungicidal compounds, anilino heterocyclic fungicidal compounds, benzimidazole type fungicidal compounds and their precursors, sulfur, copper compounds, and carbamate type fungicidal compounds for controlling fungi tolerant to benzimidazole type fungicides.

The compounds of each type are further explained below.

The ethylenebis(dithiocarbamate) type fungicidal compounds include zinc salt (Zineb), manganese salt (Maneb), salt with zinc and manganese (Mancozeb), etc.

The N-(3,5-dichlorophenyl)imide type compounds include fungicidal compounds such as N-(3,5-dichlorophenyl)-1,2-dimethylcyclopropane-1,2-dicarboximide (Procymidone), 3-(3,5-dichlorophenyl)-5-methyl-5-vinyl-1,3-oxazolidine-2,4-dione (Vinclozolin) and 3-(3,5-dichlorophenyl)-N-(1-methylethyl)-2,4-dioxo-1-imidazolidinecarboxamide (Iprodione) which are fungicidal compounds having a partial structure represented by



The phthalimide type compounds include, for example, N-(trichloromethylthio)phthalimide (Folpet) which is a compound having an N-substituted phthalimide structure, and 1,2,3,6-tetrahydro-N-(trichloromethylthio)phthalimide (Captan) and 1,2,3,6-tetrahydro-N-(1,1,2,2-tetrachloroethylthio)phthalimide (Captafol) which are compounds having an N-substituted tetrahydrophthalimide structure.

The anilide type fungicidal compounds include compounds having a 2,6-dimethylanilide structure, such as methyl N-(2,6-dimethylphenyl)-N-(methoxyacetyl)alaninate (Metalaxyl), methyl N-(2,6-dimethylphenyl)-N-(phenylacetyl)alaninate (Benalaxyl), methyl N-(2,6-dimethylphenyl)-N-(2-furanylcabonyl)alaninate (Furalaxyl), 2-chloro-N-(2,6-dimethylphenyl)-N-(tetrahydro-2-oxo-3-furanyl)acetamide (Ofurace) and 2-methoxy-N-(2-oxo-1,3-oxazolidin-3-yl)aceto-2',6'-xylylide (Oxadixyl); and compounds having a 3-chloroanilide structure, such as N-(3-chlorophenyl)-N-(tetrahydro-2-oxo-3-furanyl)cyclopropanecarboxamide (Cyprofuram).

In addition to them, there are used 2-cyano-N-[ethylaminocarbonyl]-2-(methoxyimino)acetamide (Cymoxanil) and ethyl hydrogenphosphonate (Fosetyl) or its aluminum salt, etc.

The fungicidal composition comprising the anilide type fungicidal compound, Cymoxanil or Fosetyl and the compound of the formula I is markedly effective against various plant diseases and is particularly suitably used for controlling, among these diseases, downy mildew and late blight or Phytophthora rot of cucumber, grape and potato and plant diseases caused by fungi of the genus Pythium.

The cyanopyrrole type fungicidal compounds include compounds having a 4-aryl-1H-pyrrole-3-carbonitrile structure, such as 4-(2,2-difluoro-1,3-benzodioxol-4-yl)-1H-pyrrole-3-carbonitrile (Fludioxonil).

The anilino heterocyclic fungicidal compounds include, for example, compounds having a 2-anilino-pyrimidine structure, such as 2-anilino-4,6-dimethyl-pyrimidine (Pyrimetanil) and 2-anilino-4-methyl-6-(1-propynyl)pyrimidine (Mepanipyrim); and compounds having an N-aryl-2,6-dinitroaniline structure, such as 3-chloro-N-(3-chloro-5-trifluoromethyl-2-pyridyl)- $\alpha,\alpha,\alpha$ -trifluoro-2,6-dinitro-p-toluidine (Fluazinam).

The present fungicidal composition comprising any of these cyanopyrrole type fungicidal compounds or anilino heterocyclic fungicidal compounds is suitably used for controlling gray mold (*Botrytis cinerea*).

The benzimidazole type fungicidal compounds include methyl 1-(butylcarbamoyl)-2-benzimidazole-carbamate (Benomyl), benzimidazole-2-yl carbamate (Carbendazim) and 2-(4'-thiazolyl)benzimidazole (Thiabendazole). The precursors thereof include 1,2-bis(3-ethoxycarbonyl-2-thioureido)benzene (Thiophanate) and 1,2-bis(3-methoxycarbonyl-2-thioureido)benzene (Thiophanate-methyl) which are considered to be convertible into benzimidazol-2-yl carbamate in a plant.

The copper compounds includes various inorganic salts such as chloride, oxychloride, carbonate, oxide, hydroxide, sulfate, phosphate, silicate, zinc chromate, hydrazinium sulfate, etc., and organic salts such as acetate, copper oxyquinolinate (8-hydroxyquinolinate), oxalate, bis(3-phenylsalicylate), naphthenate, linolenate, oleate, etc.

The carbamate type fungicidal compounds for controlling fungi tolerant to benzimidazole type fungicides refer to the 3,4-substituted, 3,5-substituted or 3,4,5-substituted N-phenylcarbamate compounds disclosed in Japanese Patent Unexamined Publication No. 57-171951, Japanese Patent Examined Publication Nos. 58-39802 and 4-39458, etc. More specifically, they refer to, for example, lower (number of carbon atoms: for instance, 1 to 4) alkyl phenylcarbamate compounds having substituents such as lower (number of carbon atoms: for instance, 1 to 4) alkoxy groups, lower (number of carbon atoms: for instance, 1 to 4) alkyl groups or halogen atoms at the 3- and 4- positions, 3- and 5-positions or 3-, 4- and 5-positions of the phenyl group, which are represented by Diethofencarb (isopropyl 3,4-diethoxyphenylcarbamate).

When used in a fungicidal composition, the above-mentioned carbamate type fungicidal compounds for controlling fungi tolerant to benzimidazole type fungicides are very effective in controlling not only the fungi tolerant to the benzimidazole type fungicides but also fungi sensitive to the benzimidazole type fungicides.

Plant diseases that can be controlled by the composition of the present invention blast (*Pyricularia oryzae*), Helminthosporium leaf spot (*Cochliobolus miyabeanus*) and sheath blight (*Rhizoctonia solani*) of rice; powdery mildew (*Erysiphe graminis* f. sp. *hordei*, f. sp. *tritici*), scab (*Gibberella zeae*), rust (*Puccinia striiformis*, *P. graminis*, *P. recondita*, *P. hordei*), snow blight (*Typhula* sp., *Micronectriella nivalis*), loose smut (*Ustilagotritici*, *U. nuda*), bunt (*Tilletia caries*),

eye spot (*Pseudocercospora herpotrichoides*), foot rot (*Rhizoctonia cerealis*), scald (*Rhynchosporium secalis*), leaf blight (*Septoria tritici*) and glume blotch (*Leptosphaeria nodorum*) of barley, wheat, oats and rye; melanose (*Diaporthe citri*), scab (*Elsinoe fawcetti*), penicillium rot (*Penicillium digitatum*, *P. italicum*) of citrus; blossom blight (*Sclerotinia mali*), canker (*Valsa mali*), powdery mildew (*Podosphaera leucotricha*), *Alternaria* leaf spot (*Alternaria mali*) and scab (*Venturia inaequalis*) of apple; scab (*Venturia nashicola*), black spot (*Alternaria kikuchiana*) and rust (*Gymnosporangium haraeum*) of pear; brown rot (*Sclerotinia cinerea*), scab (*Cladosporium carpophilum*) and *Phomopsis* rot (*Phomopsis* sp.) of peach; downy mildew (*Plasmopara viticola*), *Sphaceloma* scab (*Elsinoe ampelina*), ripe rot (*Glomerellacing ulata*), powdery mildew (*Uncinula necator*) and rust (*Phakopora ampelopsidis*) of grape; anthracnose (*Gloeosporium kaki*) and leaf spot (*Cercospora kaki*, *Mycosphaerella nawae*) of Japanese persimmon; cucumber downy mildew (*Pseudoperonospora cubensis*); anthracnose (*Colletotrichum lagenarium*), powdery mildew (*Sphaerotheca fuliginea*) and gummy stem blight (*Mycosphaerella melonis*) of melons and cucumbers; early blight (*Alternaria solani*), leaf mold (*Cladosporium fulvum*) and late blight (*Phytophthora infestans*) of tomato; brown spot (*Phomopsis vexans*) and powdery mildew (*Erysiphe cichoracearum*) of eggplant; leaf spot (*Alternaria japonica*) and white spot (*Cercospora brassicae*) of vegetables of Cruciferae; onion rust (*Puccinia allii*); purple stain (*Cercospora kikuchii*), *Sphaceloma* scab (*Elsinoe glycines*) and pod and stem blight (*Diaporthe phaseolorum* var. *sajae*) of soybean; kidney bean anthracnose (*Colletotrichum lindemthianum*); leaf spot (*Mycosphaerella personatum*) and leaf spot (*Cercospora arachidicola*) of peanut; powdery mildew (*Erysiphe pisi*) and downy mildew (*Peronospora pisi*) of pea; downy mildew (*Peronospora viciae*) and *Phytophthora* rot (*Phytophthora nicotianae*) of broad bean; early blight (*Alternaria solani*) and late blight (*Phytophthora infestans*) of potato; powdery mildew (*Sphaerotheca humuli*) and *Phytophthora* rot (*Phytophthora nicotianae*) of strawberry; net blister blight (*Exobasidium recticulatum*) and white scab (*Erysiphe leucospila*) of tea plant; brown spot (*Alternaria longipes*), powdery mildew (*Erysiphe cichoracearum*), anthracnose (*Colletotrichum tabacum*) and *Phytophthora* rot (*Phytophthora parasitica*) of tobacco; beet leaf spot (*Cercospora beticola*); black spot (*Diplocarpon rosae*), powdery mildew (*Sphaerotheca pannosa*) and *Phytophthora* rot (*Phytophthora megasperma*) of rose; leaf spot (*Septoria chrysanthemi indicii*) and white rust (*Puccinia horiana*) of chrysanthemum; gray mold (*Botrytis cinerea*) and stem rot (*Sclerotinia sclerotiorum*) of fruit trees such as grape and citrus and various crops such as cucumber, tomato, beans and vegetables; and plant diseases caused by *Pythium* fungi (*Pythium* sp.).

In the fungicidal composition of the present invention, although the mixing proportions of the compound of the formula I used as active ingredient and the other active ingredient(s) selected from the group of compounds (b) are not particularly limited, the mixing may be carried out so as to adjust the amount of the compound of the formula I mixed with each of the other active ingredient(s) as follows.

Next, the weight of the at least one compound selected from the group (b) and mixed with 1 part by weight of the compound of the formula I is described below.

The proportion of the ethylenebis(dithiocarbamate) compound(s) ranges usually from 0.01 to 100 parts by weight, preferably from 0.1 to 50 parts by weight, more preferably from 1 to 20 parts by weight, per part by weight of the compound of the formula I.

The mixing proportion of the N-(3,5-dichlorophenyl)imide type compound(s) is usually 0.01 to 100 parts by weight, preferably 0.1 to 50 parts by weight.

The mixing proportion of Chlorothalonil is usually 0.1 to 10000 parts by weight, preferably 1 to 1000 parts by weight.

The mixing proportion of the phthalimide type fungicidal compound(s) is usually 0.1 to 10000 parts by weight, preferably 1 to 1000 parts by weight.

The using amount of at least one compound selected from the anilide type fungicidal compounds, Cymoxanil and Fosetyl is usually 0.001 to 100 parts by weight, preferably 0.1 to 50 parts by weight.

The using amount of the cyanopyrrole type fungicidal compound(s) or anilino heterocyclic fungicidal compound(s) is usually 0.001 to 100 parts by weight, preferably 0.002 to 20 parts by weight.

The using amount of the benzimidazole type fungicidal compound(s) or precursor(s) thereof is usually 0.01 to 100 parts by weight, preferably 0.02 to 50 parts by weight.

The using amount of sulfur is usually 1 to 5000 parts by weight, preferably 5 to 1000 parts by weight.

The using amount of the copper compound(s) is usually 0.01 to 1000 parts by weight, preferably 0.1 to 100 parts by weight.

The using amount of the carbamate type fungicidal compound(s) ranges usually from 0.01 to 100 parts by weight, preferably from 0.02 to 50 parts by weight.

The fungicidal composition of the present invention is applied usually after being formulated into an emulsifiable concentrate, wettable powder, suspension, granules, dust, dry flowable concentrate, aqueous soluble concentrate, oil formulation, smoking formulation, aerosol, microcapsules or the like, by mixing with a solid carrier, liquid carrier or gaseous carrier and optionally addition of adjuvants for formulation such as surfactants, fixing agents, dispersants, stabilizers, etc.

The compounds as active ingredients are usually contained in such a formulation in a total amount of 0.1 to 99% by weight, preferably 0.2 to 90% by weight.

The solid carrier includes, for example, fine powders or granules of clays (e.g. kaolin clay, diatomaceous earth, synthetic hydrated silicon dioxide, attapulgite clay, bentonite and acid clay), talcs, other inorganic minerals (e.g. sericite, powdered quartz, powdered sulfur, activated carbon, calcium carbonate and hydrated silica), and salts for chemical fertilizers (e.g. ammonium sulfate, ammonium phosphate, ammonium nitrate, urea and ammonium chloride). The liquid carrier includes, for example, water, alcohols (e.g. methanol and ethanol), ketones (e.g. acetone, methyl ethyl ketone and cyclohexanone), aromatic hydrocarbons (e.g. benzene, toluene, xylene, ethylbenzene and methylnaphthalene), aliphatic hydrocarbons (e.g. hexane and kerosene), esters (e.g. ethyl acetate and butyl acetate), nitriles (e.g. acetonitrile and isobutyronitrile), ethers (e.g. dioxane and diisopropyl ether), acid amides (e.g. dimethylformamide and dimethylacetamide), and halogenated hydrocarbons (e.g. dichloroethane, trichloroethylene and carbon tetrachloride). The gaseous carrier includes butane gas, carbon dioxide, fluorocarbon gas, etc.

The surfactants include alkyl sulfates, alkylsulfonates, alkylarylsulfonates, alkyl aryl ethers and their polyoxyethylated products, polyethylene glycol ethers, polyhydric alcohol esters, sugar alcohol derivatives, etc.

The fixing agents and dispersants include casein, gelatin, polysaccharides (e.g. starch, gum arabic, cellulose derivatives and alginic acid), lignin derivatives, bentonite, saccharides, synthetic water-soluble polymers [e.g. poly(vinyl alcohol)s, poly(vinylpyrrolidone)s and poly(acrylic acid)s], etc. The stabilizers include PAP (acidic isopropyl phosphate), BHT (2,6-di-tert-butyl-4-methylphenol), BHA (a mixture of 2-tert-butyl-4-methoxyphenol and 3-tert-butyl-4-methoxyphenol), vegetable oils, mineral oils, fatty acids or their esters, etc.

The above-mentioned formulations are applied to plants or soil as they are or after being diluted with water or the like. When applied to soil, they may be sprinkled on the soil surface or mixed with the soil. They may be applied also by any of various methods such as seed treatment, ULV, etc.

When used as a seed-treating agent, the formulations are used by dressing of seeds, seed immersion or the like.

In addition, the formulations may be used in combination with other fungicides, insecticides, acaricides, nematocides, herbicides, seed disinfectants, fertilizers, soil conditioners, etc.

Although the applying dosage of the fungicidal composition of the present invention is varied depending on the kinds of the compounds used as active ingredients; their mixing proportions; weather conditions; type of formulation; application time, method and site; diseases to be controlled; a crop to be protected; and the like, it is usually 0.001 to 1000 g, preferably 0.1 to 100 g, per are. When the emulsifiable concentrate, wettable powder, suspension, soluble concentrate or the like is used after being diluted with water, the applying concentration thereof is 0.0001 to 1% by weight, preferably 0.001 to 0.5% by weight. The granules, dust and the like are used as they are without dilution.

For seed treatment, the fungicidal composition is used in an amount of usually 0.001 to 50 g, preferably 0.01 to 10 g, (in terms of the sum of the compounds used as active ingredients) per kg of seeds.

## EXAMPLES

The present invention is explained below in further detail with formulation examples and test examples, but the present invention is not limited to the following examples. In the following examples, parts and percents are all by weight unless otherwise specified.

### Formulation Example 1

Granules are obtained by thoroughly grinding and mixing 1 part of compound (Ia), (Ib), (Ic), (Id), (Ie), (If) or (Ig), 5 parts of an ethylenebis(dithiocarbamate) compound, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 61 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

### Formulation Example 2

Granules are obtained by thoroughly grinding and mixing 5 parts of compound (Ia), (Ib), (Ic), (Id), (Ie), (If) or (Ig), 5 parts of an ethylenebis(dithiocarbamate) compound, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 57 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

### Formulation Example 3

A dust is obtained by thoroughly grinding and mixing 0.5 part of compound (Ia), (Ib), (Ic), (Id), (Ie), (If) or (Ig), 2.5 parts of an ethylenebis(dithiocarbamate) compound, 86 parts of kaolin clay and 11 parts of talc.

Formulation Example 4

A suspension is obtained by mixing 5 parts of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 25 parts of an ethylenebis(dithiocarbamate) compound, 3 parts of polyoxyethylene sorbitan monooleate, 3 parts of carboxymethyl cellulose and 64 parts of water, followed by wet grinding to a particle size of 5 microns or less.

Formulation Example 5

A wettable powder is obtained by thoroughly grinding and mixing 10 parts of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 50 parts of an ethylenebis(dithiocarbamate) compound, 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate and 35 parts of synthetic hydrated silicon dioxide.

Formulation Example 6

An emulsifiable concentrate is obtained by mixing 5 parts of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 25 parts of an ethylenebis(dithiocarbamate) compound, 14 parts of polyoxyethylene styrylphenyl ether, 6 parts of calcium dodecylbenzenesulfonate and 50 parts of xylene.

Formulation Example 7

Granules are obtained by thoroughly grinding and mixing 1 part of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 5 parts of Procymidone, Vinclozolin or Iprodione, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 61 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 8

Granules are obtained by thoroughly grinding and mixing 5 parts of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 5 parts of Procymidone, Vinclozolin or Iprodione, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 57 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 9

A dust is obtained by thoroughly grinding and mixing 0.5 part of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 2.5 parts of Procymidone, Vinclozolin or Iprodione, 86 parts of kaolin clay and 11 parts of talc.

Formulation Example 10

A suspension is obtained by mixing 5 parts of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 25 parts of Procymidone, Vinclozolin or Iprodione, 3 parts of polyoxyethylene sorbitan monooleate, 3 parts of carboxymethyl cellulose and 64 parts of water, followed by wet grinding to a particle size of 5 microns or less.

Formulation Example 11

A wettable powder is obtained by thoroughly grinding and mixing 10 parts of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 50 parts of Procymidone, Vinclozolin or Iprodione, 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate and 35 parts of synthetic hydrated silicon dioxide.

Formulation Example 12

An emulsifiable concentrate is obtained by mixing 5 parts of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 25 parts of Procymidone, Vinclozolin or Iprodione, 14 parts of polyoxyethylene styrylphenyl ether, 6 parts of calcium dodecylbenzenesulfonate and 50 parts of xylene.

Formulation Example 13

Granules are obtained by thoroughly grinding and mixing 1 part of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 5 parts of Chlorothalonil, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of ben-



tonite and 61 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

#### Formulation Example 14

Granules are obtained by thoroughly grinding and mixing 5 parts of compound (Ia), (Ib), (Ic), (Id), (Ie), (If) or (Ig), 5 parts of Chlorothalonil compound, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 57 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

#### Formulation Example 15

A dust is obtained by thoroughly grinding and mixing 0.5 part of compound (Ia), (Ib), (Ic), (Id), (Ie), (If) or (Ig), 2.5 parts of Chlorothalonil, 86 parts of kaolin clay and 11 parts of talc.

#### Formulation Example 16

A suspension is obtained by mixing 5 parts of compound (Ia), (Ib), (Ic), (Id), (Ie), (If) or (Ig), 25 parts of Chlorothalonil, 3 parts of polyoxyethylene sorbitan monooleate, 3 parts of carboxymethyl cellulose and 64 parts of water, followed by wet grinding to a particle size of 5 microns or less.

#### Formulation Example 17

A wettable powder is obtained by thoroughly grinding and mixing 10 parts of compound (Ia), (Ib), (Ic), (Id), (Ie), (If) or (Ig), 50 parts of Chlorothalonil, 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate and 35 parts of synthetic hydrated silicon dioxide.

#### Formulation Example 18

An emulsifiable concentrate is obtained by mixing 5 parts of compound (Ia), (Ib), (Ic), (Id), (Ie), (If) or (Ig), 25 parts of Chlorothalonil compound, 14 parts of polyoxyethylene styrylphenyl ether, 6 parts of calcium dodecylbenzenesulfonate and 50 parts of xylene.

#### Formulation Example 19

Granules are obtained by thoroughly grinding and mixing 1 part of compound (Ia), (Ib), (Ic), (Id), (Ie), (If) or (Ig), 5 parts of a phthalimide type fungicidal compound, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 61 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

#### Formulation Example 20

Granules are obtained by thoroughly grinding and mixing 5 parts of compound (Ia), (Ib), (Ic), (Id), (Ie), (If) or (Ig), 5 parts of a phthalimide type fungicidal compound, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 57 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

#### Formulation Example 21

A dust is obtained by thoroughly grinding and mixing 0.5 part of compound (Ia), (Ib), (Ic), (Id), (Ie), (If) or (Ig), 2.5 parts of a phthalimide type fungicidal compound, 86 parts of kaolin clay and 11 parts of talc.

#### Formulation Example 22

A suspension is obtained by mixing 5 parts of compound (Ia), (Ib), (Ic), (Id), (Ie) or (If) (Ig), 25 parts of a phthalimide type fungicidal compound, 3 parts of polyoxyethylene sorbitan monooleate, 3 parts of carboxymethyl cellulose and 64 parts of water, followed by wet grinding to a particle size of 5 microns or less.

Formulation Example 23

A wettable powder is obtained by thoroughly grinding and mixing 10 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 50 parts of a phthalimide type fungicidal compound, 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate and 35 parts of synthetic hydrated silicon dioxide.

Formulation Example 24

An emulsifiable concentrate is obtained by mixing 5 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 25 parts of a phthalimide type fungicidal compound, 14 parts of polyoxyethylene styrylphenyl ether, 6 parts of calcium dodecylbenzenesulfonate and 50 parts of xylene.

Formulation Example 25

Granules are obtained by thoroughly grinding and mixing 1 part of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 5 parts of an anilide type fungicidal compound, Cymoxanil or Fosetyl aluminum, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 61 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 26

Granules are obtained by thoroughly grinding and mixing 5 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 5 parts of an anilide type fungicidal compound, Cymoxanil or Fosetyl aluminum, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 57 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 27

A dust is obtained by thoroughly grinding and mixing 0.5 part of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 2.5 parts of an anilide type fungicidal compound, Cymoxanil or Fosetyl aluminum, 86 parts of kaolin clay and 11 parts of talc.

Formulation Example 28

A suspension is obtained by mixing 5 parts of compound (Ia), (Ib), (Ic), (Id), (Ie) or (If) (Ig), 25 parts of an anilide type fungicidal compound, Cymoxanil or Fosetyl aluminum, 3 parts of polyoxyethylene sorbitan monooleate, 3 parts of carboxymethyl cellulose and 64 parts of water, followed by wet grinding to a particle size of 5 microns or less.

Formulation Example 29

A wettable powder is obtained by thoroughly grinding and mixing 10 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 50 parts of an anilide type fungicidal compound, Cymoxanil or Fosetyl aluminum, 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate and 35 parts of synthetic hydrated silicon dioxide.

Formulation Example 30

An emulsifiable concentrate is obtained by mixing 5 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 25 parts of an anilide type fungicidal compound, Cymoxanil or Fosetyl aluminum, 14 parts of polyoxyethylene styrylphenyl ether, 6 parts of calcium dodecylbenzenesulfonate and 50 parts of xylene.

Formulation Example 31

Granules are obtained by thoroughly grinding and mixing 1 part of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 5 parts of Fludioxonil, Pyrimetanil, Mepanipyrim or Fluazinam, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 61 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 32

Granules are obtained by thoroughly grinding and mixing 5 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 5 parts of Fludioxonil, Pyrimetanil, Mepanipyrim or Fluazinam, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 57 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 33

A dust is obtained by thoroughly grinding and mixing 0.5 part of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 2.5 parts of Fludioxonil, Pyrimetanil, Mepanipyrim or Fluazinam, 86 parts of kaolin clay and 11 parts of talc.

Formulation Example 34

A suspension is obtained by mixing 5 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie) or (If) (Ig), 25 parts of Fludioxonil, Pyrimetanil, Mepanipyrim or Fluazinam, 3 parts of polyoxyethylene sorbitan monooleate, 3 parts of carboxymethyl cellulose and 64 parts of water, followed by wet grinding to a particle size of 5 microns or less.

Formulation Example 35

A wettable powder is obtained by thoroughly grinding and mixing 10 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 50 parts of Fludioxonil, Pyrimetanil, Mepanipyrim or Fluazinam, 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate and 35 parts of synthetic hydrated silicon dioxide.

Formulation Example 36

An emulsifiable concentrate is obtained by mixing 5 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 25 parts of Fludioxonil, Pyrimetanil, Mepanipyrim or Fluazinam, 14 parts of polyoxyethylene styrylphenyl ether, 6 parts of calcium dodecylbenzenesulfonate and 50 parts of xylene.

Formulation Example 37

Granules are obtained by thoroughly grinding and mixing 1 part of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 5 parts of Benomyl, Carbendazim, Thiabendazole, Thiophanate or Thiophanate-methyl, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 61 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 38

Granules are obtained by thoroughly grinding and mixing 5 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 5 parts of Benomyl, Carbendazim, Thiabendazole, Thiophanate or Thiophanate-methyl, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 57 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 39

A dust is obtained by thoroughly grinding and mixing 0.5 part of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 2.5 parts of Benomyl, Carbendazim, Thiabendazole, Thiophanate or Thiophanate-methyl, 86 parts of kaolin clay and 11 parts of talc.

Formulation Example 40

A suspension is obtained by mixing 5 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 25 parts of Benomyl, Carbendazim, Thiabendazole, Thiophanate or Thiophanate-methyl, 3 parts of polyoxyethylene sorbitan monooleate, 3 parts of carboxymethyl cellulose and 64 parts of water, followed by wet grinding to a particle size of 5 microns or less.

Formulation Example 41

A wettable powder is obtained by thoroughly grinding and mixing 10 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 50 parts of Benomyl, Carbendazim, Thiabendazole, Thiophanate or Thiophanatemethyl, 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate and 35 parts of synthetic hydrated silicon dioxide.

Formulation Example 42

An emulsifiable concentrate is obtained by mixing 5 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 25 parts of Benomyl, Carbendazim, Thiabendazole, Thiophanate or Thiophanate-methyl, 14 parts of polyoxyethylene styrylphenyl ether, 6 parts of calcium dodecylbenzenesulfonate and 50 parts of xylene.

Formulation Example 43

Granules are obtained by thoroughly grinding and mixing 1 part of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 5 parts of sulfur, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 61 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 44

Granules are obtained by thoroughly grinding and mixing 5 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 5 parts of sulfur, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 57 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 45

A dust is obtained by thoroughly grinding and mixing 0.5 part of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 2.5 parts of sulfur, 86 parts of kaolin clay and 11 parts of talc.

Formulation Example 46

A suspension is obtained by mixing 5 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 25 parts of sulfur, 3 parts of polyoxyethylene sorbitan monooleate, 3 parts of carboxymethyl cellulose and 64 parts of water, followed by wet grinding to a particle size of 5 microns or less.

Formulation Example 47

A wettable powder is obtained by thoroughly grinding and mixing 10 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 50 parts of sulfur, 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate and 35 parts of synthetic hydrated silicon dioxide.

Formulation Example 48

An emulsifiable concentrate is obtained by mixing 5 parts of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 25 parts of sulfur, 14 parts of polyoxyethylene styrylphenyl ether, 6 parts of calcium dodecylbenzenesulfonate and 50 parts of xylene.

Formulation Example 49

Granules are obtained by thoroughly grinding and mixing 1 part of compound (Ia), (Ib), (Ic), (Ic), (Ie), (If) or (Ig), 5 parts of a copper compound, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 61 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 50

Granules are obtained by thoroughly grinding and mixing 5 parts of compound (la), (lb), (lc), (lc), (le), (lf) or (lg), 5 parts of a copper compound, 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 57 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 51

A dust is obtained by thoroughly grinding and mixing 0.5 part of compound (la), (lb), (lc), (lc), (le), (lf) or (lg), 2.5 parts of a copper compound, 86 parts of kaolin clay and 11 parts of talc.

Formulation Example 52

A suspension is obtained by mixing 5 parts of compound (la), (lb), (lc), (ld), (le) or (lf) (lg), 25 parts of a copper compound, 3 parts of polyoxyethylene sorbitan monooleate, 3 parts of carboxymethyl cellulose and 64 parts of water, followed by wet grinding to a particle size of 5 microns or less.

Formulation Example 53

A wettable powder is obtained by thoroughly grinding and mixing 10 parts of compound (la), (lb), (lc), (lc), (le), (lf) or (lg), 50 parts of a copper compound, 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate and 35 parts of synthetic hydrated silicon dioxide.

Formulation Example 54

An emulsifiable concentrate is obtained by mixing 5 parts of compound (la), (lb), (lc), (lc), (le), (lf) or (lg), 25 parts of a copper compound, 14 parts of polyoxyethylene styrylphenyl ether, 6 parts of calcium dodecylbenzenesulfonate and 50 parts of xylene.

Formulation Example 55

Granules are obtained by thoroughly grinding and mixing 5 parts of Diethofencarb, 1 part of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 61 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 56

Granules are obtained by thoroughly grinding and mixing 5 parts of Diethofencarb, 5 parts of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 1 part of synthetic hydrated silicon dioxide, 2 parts of calcium lignin sulfonate, 30 parts of bentonite and 57 parts of kaolin clay, and thoroughly kneading the resulting mixture together with water, followed by granulation and drying.

Formulation Example 57

A dust is obtained by thoroughly grinding and mixing 2.5 parts of Diethofencarb compound, 0.5 part of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 86 parts of kaolin clay and 11 parts of talc.

Formulation Example 58

A suspension is obtained by mixing 25 parts of Diethofencarb compound, 5 parts of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 3 parts of polyoxyethylene sorbitan monooleate, 3 parts of carboxymethyl cellulose and 64 parts of water, followed by wet grinding to a particle size of 5 microns or less.

## Formulation Example 59

A wettable powder is obtained by thoroughly grinding and mixing 50 parts of Diethofencarb compound, 10 parts of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate and 35 parts of synthetic hydrated silicon dioxide.

## Formulation Example 60

An emulsifiable concentrate is obtained by mixing 25 parts of Diethofencarb compound, 5 parts of compound (la), (lb), (lc), (ld), (le), (lf) or (lg), 14 parts of polyoxyethylene styrylphenyl ether, 6 parts of calcium dodecylbenzenesulfonate and 50 parts of xylene.

## Test Example 1

A tomato plant (cultivar: Ponterosa) was planted in each pot and was sprayed with a sufficient volume of a dilution with a predetermined concentration of a wettable powder obtained according to Formulation Example 5, after the plant put out a third leaf. After air-drying of the liquid chemical, the surfaces of the leaves were inoculated with a suspension of spores ( $5 \times 10^4$  spores/ml) of tomato late blight fungus (*Phytophthora infestans*) by spraying. The plant was grown at 24°C for 1 week, after which the infection rate (%) was investigated.

The results are shown in Table 1.

Table 1

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (la)	10	25
Compound (lb)	50	25
Compound (lc)	10	25
Compound (ld)	10	25
Compound (le)	10	25
Mancozeb	50	25
Compound (la) + Mancozeb	10 + 50	0
Compound (lb) + Mancozeb	50 + 50	0
Compound (lc) + Mancozeb	10 + 50	0
Compound (ld) + Mancozeb	10 + 50	0
Compound (le) + Mancozeb	10 + 50	0

## Test Example 2

A grape plant (cultivar: Berry A) was planted in each pot and was sprayed with a sufficient volume of a dilution with a predetermined concentration of a wettable powder obtained according to Formulation Example 5, after the plant put out a fourth leaf. After air-drying of the liquid chemical, the surfaces of the leaves were inoculated with a suspension of spores ( $2 \times 10^5$  spores/ml) of grape downy mildew fungus (*Plasmopara viticola*) by spraying. The plant was grown at 24°C for 1 week, after which the infection rate (%) was investigated.

The results are shown in Table 2.

Table 2

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	10	30
Compound (Ib)	50	25
Compound (Ic)	10	25
Compound (Id)	10	25
Compound (Ie)	10	25
Mancozeb	50	25
Compound (Ia) + Mancozeb	10 + 50	0
Compound (Ib) + Mancozeb	50 + 50	0
Compound (Ic) + Mancozeb	10 + 50	0
Compound (Id) + Mancozeb	10 + 50	0
Compound (Ie) + Mancozeb	10 + 50	0

### Test Example 3

Each of plastic pots was filled with sandy loam and sown with cucumber (Sagamihanjiro), followed by growing in a greenhouse for 14 days. A wettable powder of test agents prepared according to Formulation Example 11 was diluted to a predetermined concentration with water, and foliage application of the dilution was carried out so as to adhere the dilution sufficiently to the surfaces of leaves of the grown cucumber young seedling. After air-drying, gray mold fungus was inoculated on the surfaces of leaves of the young seedling, and the young seedling was grown in the dark at 10°C and a high humidity for 4 days, after which the infection rate (%) was investigated.

The results are shown in Table 3.

Table 3

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	10	30
Compound (Ib)	200	20
Compound (Ic)	200	20
Compound (Id)	200	20
Compound (Ie)	20	25
Procymidone	10	25
Compound (Ia) + Procymidone	10 + 10	0
Compound (Ib) + Procymidone	200 + 10	0
Compound (Ic) + Procymidone	200 + 10	0
Compound (Id) + Procymidone	200 + 10	0
Compound (Ie) + Procymidone	20 + 10	0

## Test Example 4

Each of plastic pots was filled with sandy loam and sown with cucumber (Sagamihanjiro), followed by growing in a greenhouse for 14 days. Gray mold fungus was inoculated on the surfaces of leaves of the grown cucumber young seedling, and the young seedling was grown in the dark at 10°C and a high humidity for 2 days and then at a temperature of 23°C for 3 days. A wettable powder of test agents prepared according to Formulation Example 11 was diluted to a predetermined concentration with water, and foliage application of the dilution was carried out so as to adhere the dilution sufficiently to the surfaces of leaves of the cucumber young seedling. After the spraying, the young seedling was grown in the dark at 10°C and a high humidity for 3 days, and then the infection rate (%) was investigated.

The results are shown in Table 4.

Table 4

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	10	30
Compound (Ib)	200	20
Compound (Ic)	200	20
Compound (Id)	200	20
Compound (Ie)	20	25
Procymidone	10	25
Compound (Ia) + Procymidone	10 + 10	0
Compound (Ib) + Procymidone	200 + 10	0
Compound (Ic) + Procymidone	200 + 10	0
Compound (Id) + Procymidone	200 + 10	0
Compound (Ie) + Procymidone	20 + 10	0

## Test Example 5

Each of plastic pots was filled with sandy loam and sown with tomato (Ponterosa), followed by growing in a greenhouse for 20 days. A wettable powder of test agents prepared according to Formulation Example 17 was diluted to a predetermined concentration with water, and foliage application of the dilution was carried out so as to adhere the dilution sufficiently to the surfaces of leaves of the tomato young seedling which had put out two true leaves. Then, the young seedling was inoculated with a suspension of zoosporangia of tomato late blight fungus by spraying. After the inoculation, the young seedling was allowed to stand overnight at 23°C and a high humidity and then grown in a greenhouse for 4 days, after which the infection rate (%) was investigated.



The results are shown in Table 5.

Table 5

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	50	25
Compound (Ib)	40	20
Compound (Ic)	40	20
Compound (Id)	40	20
Compound (Ie)	10	20
Chlorothalonil	40	20
Compound (Ia) + Chlorothalonil	50 + 40	0
Compound (Ib) + Chlorothalonil	40 + 40	0
Compound (Ic) + Chlorothalonil	40 + 40	0
Compound (Id) + Chlorothalonil	40 + 40	0
Compound (Ie) + Chlorothalonil	10 + 40	0

#### Test Example 6

Each of plastic pots was filled with sandy loam and sown with grape (Berry A), followed by growing in a greenhouse for 40 days. A suspension of test agents prepared according to Formulation Example 16 were diluted to a predetermined concentration with water, and foliage application of the dilution was carried out so as to adhere the dilution sufficiently to the surfaces of leaves of the grape young seedling which had put out three true leaves. Then, the young seedling was inoculated with a suspension of zoosporangia of grape downy mildew fungus by spraying. After the inoculation, the young seedling was allowed to stand overnight at 23°C and a high humidity and then grown in a greenhouse for 7 days, after which the infection rate (%) was investigated.

The results are shown in Table 6.

Table 6

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	5	20
Compound (Ib)	5	20
Compound (Ic)	5	25
Compound (Id)	5	25
Compound (Ie)	1	25
Chlorothalonil	30	20
Compound (Ia) + Chlorothalonil	5 + 30	0
Compound (Ib) + Chlorothalonil	5 + 30	0
Compound (Ic) + Chlorothalonil	5 + 30	0
Compound (Id) + Chlorothalonil	5 + 30	0
Compound (Ie) + Chlorothalonil	1 + 30	0

#### Test Example 7

Each of plastic pots was filled with sandy loam and sown with grape (Berry A), followed by growing in a greenhouse for 40 days. A wettable powder prepared of test agents according to Formulation Example 23 was diluted to a predetermined concentration with water, and foliage application of the dilution was carried out so as to adhere the dilution sufficiently to the surfaces of leaves of the grape young seedling which had put out three true leaves. Then, the young seedling was inoculated with a suspension of zoosporeangia of grape downy mildew fungus by spraying. After the inoculation, the young seedling was allowed to stand overnight at 23°C and a high humidity and then grown in a greenhouse for 7 days, after which the infection rate (%) was investigated.

The results are shown in Table 7.

Table 7

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	10	20
Compound (Ib)	10	20
Compound (Ic)	10	25
Compound (Id)	10	20
Compound (Ie)	2	20
Folpet	200	20
Compound (Ia) + Folpet	10+200	0
Compound (Ib) + Folpet	10+200	0
Compound (Ic) + Folpet	10+200	0
Compound (Id) + Folpet	10+200	0
Compound (Ie) + Folpet	2+200	0

#### Test Example 8

Each of plastic pots was filled with sandy loam and sown with apple (Fuji), followed by growing in a greenhouse for 20 days. A suspension of test agents prepared according to Formulation Example 22 was diluted to a predetermined concentration with water, and foliage application of the dilution was carried out so as to adhere the dilution sufficiently to the surfaces of leaves of the apple young seedling which had put out a fourth or fifth true leaf. Then, the young seedling was inoculated with a suspension of spores of apple scab fungus by spraying. After the inoculation, the young seedling was allowed to stand in the dark at 15°C and a high humidity for 4 days and then grown under illumination for 15 days, after which the infection rate (%) was investigated.

The results are shown in Table 8.

Table 8

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	0.8	20
Compound (Ib)	2	25
Compound (Ic)	5	25
Compound (Id)	5	25
Compound (Ie)	0.5	20
Captan	300	20
Compound (Ia) + Captan	0.8 + 300	0
Compound (Ib) + Captan	2 + 300	0
Compound (Ic) + Captan	5 + 300	0
Compound (Id) + Captan	5 + 300	0
Compound (Ie) + Captan	0.5 + 300	0

#### Test Example 9

Each of plastic pots was filled with sandy loam and sown with grape (Berry A), followed by growing in a greenhouse for 40 days. A wettable powder of test agents prepared according to Formulation Example 29 was diluted to a predetermined concentration with water, and foliage application of the dilution was carried out so as to adhere the dilution sufficiently to the surfaces of leaves of the grape young seedling which had put out three true leaves. Then, the young seedling was inoculated with a suspension of zoospores of grape downy mildew fungus by spraying. After the inoculation, the young seedling was allowed to stand overnight at 23°C and a high humidity and then grown in a greenhouse for 7 days, after which the infection rate (%) was investigated.

The results are shown in Table 9.

Table 9

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	20	25
Compound (Ib)	40	20
Compound (Ic)	100	20
Compound (Id)	40	20
Compound (Ie)	10	25
Cymoxanil	40	20
Metalaxyl	1	20
Benalaxyl	1	25
	1	25
Oxadixyl	5	20
Fosetyl aluminum	100	25
Compound (Ia) + Cymoxanil	20+40	0
Compound (Ib) + Cymoxanil	40+40	0
Compound (Ic) + Cymoxanil	100+40	0
Compound (Id) + Cymoxanil	40+40	0
Compound (Ie) + Cymoxanil	10+40	0
Compound (Ia) + Metalaxyl	40+ 1	0
Compound (Ie) + Metalaxyl	10+ 1	0
Compound (Ie) + Benalaxyl	40+ 1	0
Compound (Ie) + Ofurace	10+ 1	0
Compound (Ie) + Oxadixyl	10+ 5	0
Compound (Ia) + Fosetyl aluminum	40+100	0
Compound (Ie) + Fosetyl aluminum	10+100	0

#### Test Example 10

Each of plastic pots was filled with sandy loam and sown with tomato (Ponterosa), followed by growing in a greenhouse for 20 days. A wettable powder of test agents prepared according to Formulation Example 35 was diluted to a predetermined concentration with water, and foliage application of the dilution was carried out so as to adhere the dilution sufficiently to the surfaces of leaves of the tomato young seedling which had put out two true leaves. Then, the young seedling was inoculated with a suspension of zoosporangia of tomato late blight fungus by spraying. After the inoculation, the young seedling was allowed to stand overnight at 23°C and a high humidity and then grown in a greenhouse for 4 days, after which the infection rate (%) was investigated.

# EP 0 741 970 A1

The results are shown in Table 10.

Table 10

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	100	25
Compound (Ib)	40	20
Compound (Ic)	40	20
Compound (Id)	40	20
Compound (Ie)	5	25
Metalaxyl	5	20
Compound (Ia) + Metalaxyl	100 + 5	0
Compound (Ib) + Metalaxyl	40 + 5	0
Compound (Ic) + Metalaxyl	40 + 5	0
Compound (Id) + Metalaxyl	40 + 5	0
Compound (Ie) + Metalaxyl	5 + 5	0

## Test Example 11

Each of plastic pots was filled with sandy loam and sown with cucumber (Sagamihanjiro), followed by growing in a greenhouse for 14 days. A wettable powder of test agents prepared according to Formulation Example 35 was diluted to a predetermined concentration with water, and foliage application of the dilution was carried out so as to adhere the dilution sufficiently to the surfaces of leaves of the grown cucumber young seedling. After air-drying, gray mold fungus was inoculated on the surfaces of leaves of the young seedling, and the young seedling was grown in the dark at 10°C and a high humidity for 4 days, after which the infection rate (%) was investigated.

The results are shown in Table 11.

Table 11

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	10	30
Compound (Ib)	200	20
Compound (Ic)	200	20
Compound (Id)	200	20
Compound (Ie)	20	25
Fludioxonil	0.5	20
Pyrimetanil	10	20
Mepanipyrim	5	20
Fluazinam	1	25
Compound (Ia) + Fludioxonil	10+0.5	0
Compound (Id) + Fludioxonil	200+0.5	0
Compound (Ie) + Fludioxonil	20+0.5	0
Compound (Ia) + Pyrimetanil	10+10	0
Compound (Ib) + Pyrimetanil	200+10	0
Compound (Ie) + Pyrimetanil	20+10	0
Compound (Ia) + Mepanipyrim	10+ 5	0
Compound (Ic) + Mepanipyrim	200+ 5	0
Compound (Ie) + Mepanipyrim	20+ 5	0
Compound (Ia) + Fluazinam	10+ 1	0
Compound (Ie) + Fluazinam	20+ 1	0

#### Test Example 12

Each of plastic pots was filled with sandy loam and sown with cucumber (Sagamihanjiro), followed by growing in a greenhouse for 14 days. A wettable powder of test agents prepared according to Formulation Example 41 was diluted to a predetermined concentration with water, and foliage application of the dilution was carried out so as to adhere the dilution sufficiently to the surfaces of leaves of the grown cucumber young seedling. After air-drying, each of gray mold fungi sensitive and tolerant, respectively, to benzimidazole type fungicidal compounds was inoculated on the surfaces of leaves of the young seedling, and the young seedling was grown in the dark at 10°C and a high humidity for 4 days, after which the infection rate (%) was investigated.

The results are shown in Table 12.

Table 12

Agent tested	Amount of agent applied (ppm)	Infection rate (%)	
		Sensitive fungus	Resistant fungus
Compound (Ia)	10	30	30
Compound (Ib)	200	20	20
Compound (Ic)	200	20	20
Compound (Id)	200	20	20
Compound (Ie)	20	25	25
Benomyl	10	20	100
Carbendazim	5	20	100
Thiabendazole	30	25	100
Thiophanate-methyl	10	25	100
Compound (Ia) + Benomyl	10+10	0	0
Compound (Ia) + Carbendazim	10+ 5	0	0
Compound (Ia) + Thiabendazole	10+30	0	0
Compound (Ia) + Thiophanate-methyl	10+10	0	0
Compound (Ib) + Benomyl	200+10	0	0
Compound (Ic) + Carbendazim	200+ 5	0	0
Compound (Id) + Thiophanate-methyl	200+10	0	0
Compound (Ie) + Benomyl	20+10	0	0
Compound (Ie) + Carbendazim	20+ 5	0	0
Compound (Ie) + Thiabendazole	20+30	0	0
Compound (Ie) + Thiophanate-methyl	20+10	0	0

#### Test Example 13

Each of plastic pots was filled with sandy loam and sown with apple (Fuji), followed by growing in a greenhouse for 20 days. A suspension of test agents prepared according to Formulation Example 40 was diluted to a predetermined concentration with water, and foliage application of the dilution was carried out so as to adhere the dilution sufficiently to the surfaces of leaves of the apple young seedling which had put out a fourth or fifth true leaf. Then, the young seedling was inoculated with a suspension of spores of apple scab fungus by spraying. After the inoculation, the young seedling was allowed to stand in the dark at 15°C and a high humidity for 4 days and then grown under illumination for 15 days, after which the infection rate (%) was investigated.



The results are shown in Table 13.

Table 13

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	1	20
Compound (Ib)	1	25
Compound (Ic)	1	30
Compound (Id)	5	20
Compound (Ie)	0.5	20
Benomyl	20	25
Carbendazim	20	25
Thiophanate-methyl	10	20
Compound (Ia) + Benomyl	1 + 20	0
Compound (Ia) + Carbendazim	1 + 20	0
Compound (Ib) + Thiophanate-methyl	1 + 10	0
Compound (Ic) + Thiophanate-methyl	1 + 10	0
Compound (Id) + Thiophanate-methyl	5 + 10	0
Compound (Ie) + Benomyl	0.5 + 20	0
Compound (Ie) + Carbendazim	0.5 + 20	0
Compound (Ie) + Thiophanate-methyl	0.5 + 10	0

## Test Example 14

Each of plastic pots was filled with sandy loam and planted with a grape (Berry A) plant, and a wettable powder of test agents prepared according to Formulation Example 47 was diluted to a predetermined concentration with water and sprayed on the plant so as to adhere to the surfaces of leaves of the plant, after the plant put out a fourth leaf. After air-drying, the surfaces of the leaves were inoculated with a suspension of spores ( $2 \times 10^5$  spores/ml) of grape powdery mildew fungus (*Uncinula necator*) by spraying. After the inoculation, the plant was grown at 24°C for 3 weeks and then the infection rate (%) was investigated.

The results are shown in Table 14.

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	1	20
Compound (Ib)	1	20
Compound (Ic)	1	20
Compound (Id)	1	25
Compound (Ie)	1	25
Sulfur	500	20
Compound (Ia) + Sulfur	1+500	0
Compound (Ib) + Sulfur	1+500	0
Compound (Ic) + Sulfur	1+500	0
Compound (Id) + Sulfur	1+500	0
Compound (Ie) + Sulfur	1+500	0

#### Test Example 15

A tomato plant (cultivar: Ponterosa) was planted in each pot and was sprayed with a sufficient volume of a dilution with a predetermined concentration of a wettable powder obtained according to Formulation Example 53, after the plant put out a third leaf. After air-drying of the spray mix, the surfaces of the leaves were inoculated with a suspension of spores ( $5 \times 10^4$  spores/ml) of tomato late blight fungus (*Phytophthora infestans*) by spraying. After the inoculation, the plant was grown at 24°C for 1 week and then the infection rate (%) was investigated.

The results are shown in Table 15.

Table 15

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	10	25
Compound (Ib)	50	25
Compound (Ic)	10	25
Compound (Id)	10	25
Compound (Ie)	10	25
Copper oxychloride	500	25
Copper oxyquinolinolate	50	25
Compound (Ia) + Copper oxychloride	10 + 500	0
Compound (Ia) + Copper oxyquinolinolate	10 + 500	0
Compound (Ib) + Copper oxychloride	50 + 500	0
Compound (Ic) + Copper oxychloride	10 + 500	0
Compound (Id) + Copper oxychloride	10 + 500	0
Compound (Ie) + Copper oxychloride	10 + 500	0
Compound (Ie) + Copper oxyquinolinolate	10 + 50	0

#### Test Example 16

Each of plastic pots was filled with sandy loam and planted with a grape (Berry A) plant, and a wettable powder of test agents prepared according to Formulation Example 53 was diluted to a predetermined concentration and sprayed on the plant so as to adhere to the surfaces of leaves of the plant, after the plant put out a fourth leaf. After air-drying, the surfaces of the leaves were inoculated with a suspension of spores ( $2 \times 10^5$  spores/ml) of grape powdery mildew fungus (*Uncinula necator*) by spraying. After the inoculation, the plant was grown at 24°C for 3 weeks and then the infection rate (%) was investigated.

The results are shown in Table 16.

Table 16

Agent tested	Amount of agent applied (ppm)	Infection rate (%)
Compound (Ia)	10	30
Compound (Ib)	50	25
Compound (Ic)	10	25
Compound (Id)	10	25
Compound (Ie)	10	25
Copper oxychloride	500	30
Copper oxyquinolinolate	200	25
Compound (Ia) + Copper oxychloride	10 + 500	0
Compound (Ia) + Copper oxyquinolinolate	10 + 200	0
Compound (Ib) + Copper oxychloride	50 + 500	0
Compound (Ic) + Copper oxychloride	10 + 500	0
Compound (Id) + Copper oxychloride	10 + 500	0
Compound (Ie) + Copper oxychloride	10 + 500	0
Compound (Ie) + Copper oxyquinolinolate	10 + 200	0

#### Test Example 17

Each of plastic pots was filled with sandy loam and sown with cucumber (Sagamihanjiro), followed by growing in a greenhouse for 14 days. A wettable powder of test agents prepared according to Formulation Example 59 was diluted to a predetermined concentration with water, and foliage application of the dilution was carried out so as to adhere the dilution sufficiently to the surfaces of leaves of the grown cucumber young seedling. After air-drying, each of gray mold fungi sensitive and tolerant, respectively, to benzimidazole type fungicidal compounds was inoculated on the surfaces of leaves of the young seedling, and the young seedling was grown in the dark at 10°C and a high humidity for 4 days, after which the infection rate (%) was investigated.

The results are shown in Table 17.

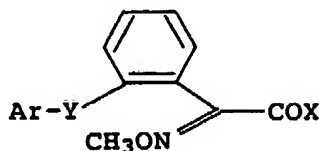
Table 17

Agent tested	Amount of agent applied (ppm)	Infection rate (%)	
		Sensitive fungus	Resistant fungus
Diethofencarb	10	100	20
Compound (Ia)	10	30	30
Compound (Ib)	200	20	20
Compound (Ic)	200	20	20
Compound (Id)	200	20	20
Compound (Ie)	20	25	25
Diethofencarb + Compound (Ia)	10 + 10	0	0
Diethofencarb + Compound (Ib)	10 + 200	0	0
Diethofencarb + Compound (Ic)	10 + 200	0	0
Diethofencarb + Compound (Id)	10 + 200	0	0
Diethofencarb + Compound (Ie)	10 + 20	0	0

### Claims

1. A fungicidal composition characterized by containing as active ingredients

(a) a compound represented by the formula I:



(wherein Ar is a substituted or unsubstituted phenyl group, Y is an oxygen atom, an oxymethylene group or a methyleneoxy group, X is  $\text{NR}_1\text{R}_2$  or  $\text{OR}_3$ , and  $\text{R}_1$ ,  $\text{R}_2$  and  $\text{R}_3$ , which may be the same or different, are hydrogen atoms or alkyl groups having 1 to 4 carbon atoms), and

(b) at least one compound selected from the group consisting of ethylenebis(dithiocarbamate) type fungicidal compounds, N-(3,5-dichlorophenyl)imide type fungicidal compounds, Chlorothalonil, phthalimide type fungicidal compounds, anilide type fungicidal compounds, Cymoxanil, Fosetyl, cyanopyrrole type fungicidal compounds, anilino type heterocyclic fungicidal compounds, benzimidazole type fungicidal compounds and their precursors, sulfur, copper compounds, and carbamate type fungicidal compounds for controlling fungi tolerant to benzimidazole type fungicides.

2. A fungicidal composition according to claim 1, wherein the compound of the general formula I is

- (Ia) methyl  $\alpha$ -methoxyimino-2-[(2-methylphenoxy)methyl]phenylacetate
- (Ib) N-methyl- $\alpha$ -methoxyimino-2-[(4-chloro-2-methylphenoxy)methyl]phenylacetamide,
- (Ic) N-methyl- $\alpha$ -methoxyimino-2-[(3-chlorophenoxy)methyl]phenylacetamide,
- (Id) N-methyl- $\alpha$ -methoxyimino-2-[(2,4-dichlorophenoxy)methyl]phenylacetamide,
- (Ie) N-methyl- $\alpha$ -methoxyimino-2-[(2,5-dimethylphenoxy)methyl]phenylacetamide,
- (If) N-methyl- $\alpha$ -methoxyimino-2-[(3-trifluoromethylphenoxy)methyl]phenylacetamide, or

(lg) N-methyl- $\alpha$ -methoxyimino-2-phenoxyphenylacetamide.

3. A fungicidal composition according to claim 1 or 2, wherein the at least one compound selected from the group of compounds (b) is an ethylenebis(dithiocarbamate) type fungicidal compound.
- 5 4. A fungicidal composition according to claim 1 or 2, wherein the at least one compound selected from the group of compounds (b) is an N-(3,5-dichlorophenyl)imide type fungicidal compound.
- 10 5. A fungicidal composition according to claim 1 or 2, wherein the at least one compound selected from the group of compounds (b) is Chlorothalonil.
6. A fungicidal composition according to claim 1 or 2, wherein the at least one compound selected from the group of compounds (b) is a phthalimide type fungicidal compound.
- 15 7. A fungicidal composition according to claim 1 or 2, wherein the at least one compound selected from the group of compounds (b) is an anilide type fungicidal compound, Cymoxanil or Fosetyl.
8. A fungicidal composition according to claim 1 or 2, wherein the at least one compound selected from the group of compounds (b) is a cyanopyrrole type fungicidal compound or an anilino heterocyclic fungicidal compound.
- 20 9. A fungicidal composition according to claim 1 or 2, wherein the at least one compound selected from the group of compounds (b) is a benzimidazole type fungicidal compound or a precursor thereof.
10. A fungicidal composition according to claim 1 or 2, wherein the at least one compound selected from the group of compounds (b) is sulfur.
- 25 11. A fungicidal composition according to claim 1 or 2, wherein the at least one compound selected from the group of compounds (b) is a copper compound.
- 30 12. A fungicidal composition according to claim 1 or 2, wherein the at least one compound selected from the group of compounds (b) is a carbamate type fungicidal compound for controlling fungi tolerant to benzimidazole type fungicides.
- 35 13. A fungicidal composition according to claim 3, wherein the ethylenebis(dithiocarbamate) type fungicidal compound is zinc salt (Zineb), manganese salt (Maneb), or a salt with zinc and manganese (Mancozeb).
- 40 14. A fungicidal composition according to claim 4, wherein the N-(3,5-dichlorophenyl)imide type fungicidal compound is 2-dimethylcyclopropane-1,2-dicarboxyimide (Procymidone), 3-(3,5-dichlorophenyl)-5-methyl-5-vinyl-1,3-oxazolidine-2,4-dione (Vinclozolin) or 3-(3,5-dichlorophenyl)-N-(1-methylethyl)-2,4-dioxo-1-imidazolidinecarboxamide (Iprodione).
- 45 15. A fungicidal composition according to claim 6, wherein the phthalimide type fungicidal compound is N-(trichloromethylthio)phthalimide (Folpet), 1,2,3,6-tetrahydro-N-(trichloromethylthio)phthalimide (Captan) or 1,2,3,6-tetrahydro-N-(1,1,2,2-tetrachloroethylthio)phthalimide (Captafol).
- 50 16. A fungicidal composition according to claim 7, wherein the anilide type fungicidal compound is methyl N-(2,6-dimethylphenyl)-N-(methoxyacetyl)alaninate (Metalaxyl), methyl N-(2,6-dimethylphenyl)-N-(phenylacetyl)alaninate (Benalazyl), methyl N-(2,6-dimethylphenyl)-N-(2-furanylcarbonyl)alaninate (Furalaxyl), 2-chloro-N-(2,6-dimethylphenyl)-N-(tetrahydro-2-oxo-3-furanyl)acetamide (Ofurace), 2-methoxy-N-(2-oxo-1,3-oxazolidin-3-yl)aceto-2',6'-xylylide (Oxadixyl) or N-(3-chlorophenyl)-N-(tetrahydro-2-oxo-3-furanyl)cyclopropanecarboxamide (Cyprofuram).
- 55 17. A fungicidal composition according to claim 8, wherein the cyanopyrrole type fungicidal compound is 4-(2,2-difluoro-1,3-benzodioxol-4-yl)-1H-pyrrole-3-carbonitrile (Fludioxonil), and the anilino heterocyclic fungicidal compound is 2-anilino-4,6-dimethylpyrimidine (Pyrimetanil), 2-anilino-4-methyl-6-(1-propynyl)pyrimidine (Mepanipyrim), or 3-chloro-N-(3-chloro-5-trifluoromethyl-2-pyridyl)- $\alpha,\alpha,\alpha$ -trifluoro-2,6-dinitro-p-toluidine (Fluazinam).
18. A fungicidal composition according to claim 9, wherein the benzimidazole type fungicidal compound or precursor thereof is methyl 1-(butylcarbamoyl)-2-benzimidazolecarbamate (Benomyl), benzimidazole-2-yl carbamate

**EP 0 741 970 A1**

(Carbendazim), 2-(4'-thiazolyl)benzimidazole (Thiabendazole), 1,2-bis(3-ethoxycarbonyl-2-thioureido)benzene (Thiophanate) or 1,2-bis(3-methoxycarbonyl-2-thioureido)benzene (Thiophanate-methyl).

5 19. A fungicidal composition according to claim 11, wherein the copper compound is chloride, oxychloride, carbonate, oxide, hydroxide, sulfate, phosphate, silicate, zinc chromate, hydrazinium sulfate, acetate, 8-hydroxyquinolate, oxalate, bis(3-phenylsalicylate), naphthenate, linolenate or oleate of copper.

10 20. A fungicidal composition according to claim 12, wherein the carbamate type fungicidal compound for controlling fungi tolerant to benzimidazole type fungicides is Diethofencarb (isopropyl 3,4-diethoxyphenylcarbamate).

21. A method for controlling plant diseases which uses a fungicidal composition set forth in any of claims 1 to 20.

15

20

25

30

35

40

45

50

55

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP94/02023

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> Int. Cl. <sup>6</sup> A01N37/50, A01N37/22, A01N37/32, A01N37/34, A01N43/08, A01N43/36, A01N43/40, A01N43/50, A01N43/54, A01N43/76 According to International Patent Classification (IPC) or to both national classification and IPC		
<b>B. FIELDS SEARCHED</b> Minimum documentation searched (classification system followed by classification symbols) Int. Cl. <sup>6</sup> A01N37/22, A01N37/32, A01N37/34, A01N37/50, A01N43/08, A01N43/36, A01N43/40, A01N43/50, A01N43/54, A01N43/76 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)		
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP, A, 63-23852 (BASF AG.), February 1, 1988 (01. 02. 88), &EP, A1, 253213	1-4, 6-9, 11, 13-16 18, 19, 21
Y		5, 10, 12 17, 20
X	JP, A, 63-30463 (Imperial Chemical Industries PLC.), February 9, 1988 (09. 02. 88), &EP, A2, 254426	1-7, 9, 11, 13-16 18, 19, 21
Y		8, 10, 12 17, 20
Y	JP, A, 3-246268 (Shionogi & Co., Ltd.), November 1, 1991 (01. 11. 91), &EP, A2, 398692	1-21
Y	JP, A, 4-288045 (BASF AG.), October 13, 1992 (13. 10. 92), &EP, A1, 477631	1-21
<input checked="" type="checkbox"/> Further documents are listed in the continuation of Box C. <input type="checkbox"/> See patent family number.		
* Special categories of cited documents: "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international filing date "L" document which may have priority claim(s) or which is cited to establish the publication date of another document or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than its priority date		
"X" document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "Y" document of particular relevance; the claimed invention appears to be novel or inventive or to be considered to involve an inventive step when the document is taken into account "Z" document of particular relevance; the claimed invention appears to be novel or inventive or to be considered to involve an inventive step when the document is taken into account or when other such documents, each establishing being relevant to a point related to the art "A" document member of the same patent family		
Date of the actual completion of the international search		Date of mailing of the international search report
February 22, 1995 (22. 02. 95)		March 14, 1995 (14. 03. 95)
Name and mailing address of the ISA/ Japanese Patent Office		Authorized officer
Facsimile No.		Telephone No.

Form PCT/ISA/210 (second sheet) (July 1992)



## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP94/02023

## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP, A, 5-194111 (BASF AG.), August 3, 1993 (03. 08. 93), &EP, A1, 524496	1-21
Y	JP, A, 4-182461 (Shionogi & Co., Ltd.), June 30, 1992 (30. 06. 92), (Family: none)	1-21
Y	JP, A, 59-204165 (Sumitomo Chemical Co., Ltd.), November 19, 1984 (19. 11. 84), (Family: none)	1, 2, 12, 20, 21
Y	JP, A, 4-9307 (Kumiai Chemical Industry Co., Ltd.), January 14, 1992 (14. 01. 92), (Family: none)	1, 2, 10, 21
Y	JP, A, 4-266805 (CIBA-Geigy AG.), September 22, 1992 (22. 09. 92), &EP, A1, 488945	1, 2, 8, 17, 21
Y	JP, A, 62-106084 (Kumiai Chemical Industry Co., Ltd.), May 16, 1987 (16. 05. 87), (Family: none)	1, 2, 8, 17, 21

Form PCT/ISA/210 (continuation of second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP94/02023

Box III TEXT OF THE ABSTRACT (Continuation of item 5 of the first sheet)

A(Continuation). CLASSIFICATION OF SUBJECT MATTER

A01N43/78, A01N47/10, A01N47/30, A01N53/12, A01N55/02,  
A01N59/02

B(Continuation). FIELDS SEARCHED

A01N43/78, A01N47/10, A01N47/30, A01N53/12, A01N55/02,  
A01N59/02